

Measurement of rotational levels of the homonuclear helium dimer cation by extrapolation of Rydberg series

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RE03, June 24, 9:04am, 2015 McPherson Lab

Introduction: the He_2^+ molecule

- three-electron system, candidate for highly accurate *ab initio* calculations

Introduction: the He_2^+ molecule

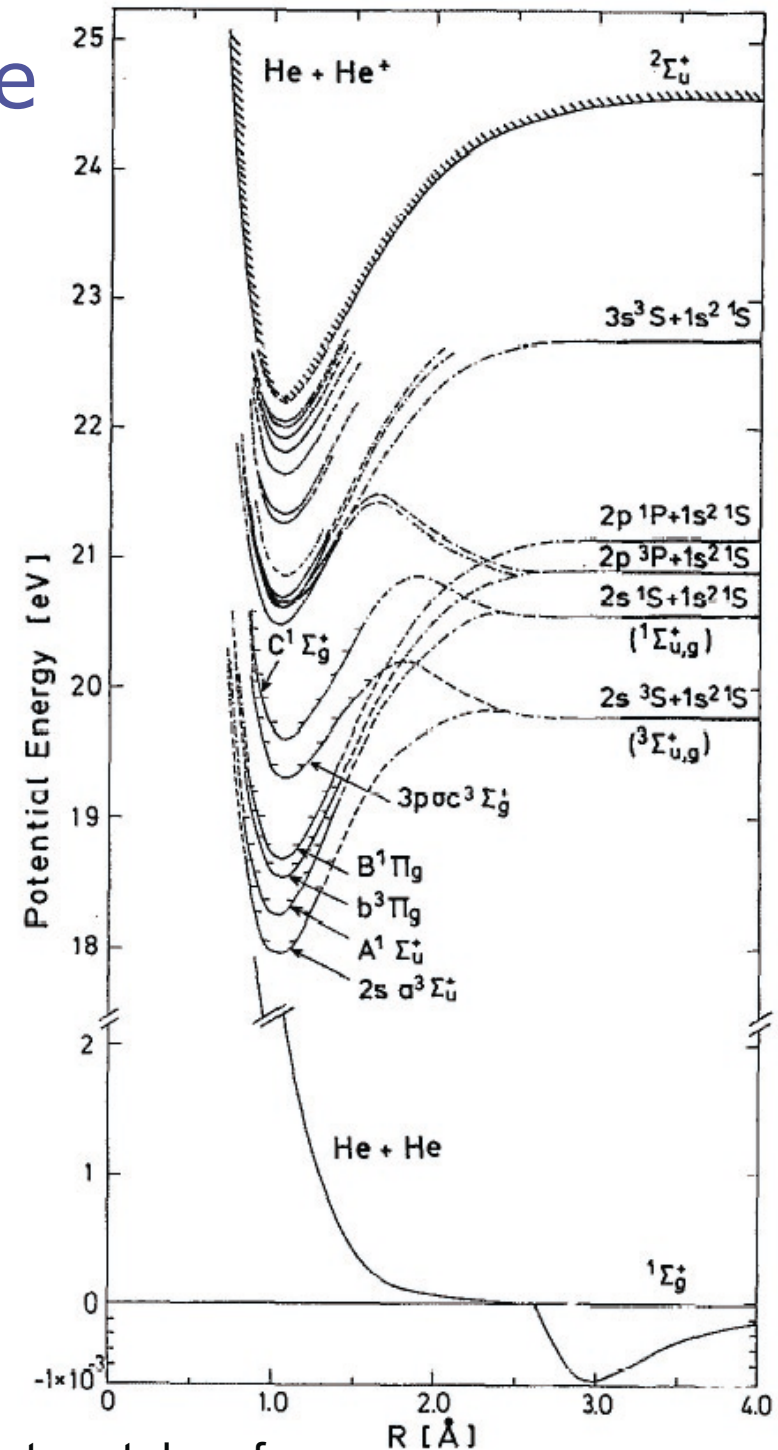
- three-electron system, candidate for highly accurate *ab initio* calculations
- not much experimental data available:
 - 9 rovibrational $v=0 \rightarrow 1$ transitions in $^3\text{He}^4\text{He}^+$ (uncertainty 18 MHz) [1]
 - 7 rovibronic $X \rightarrow A$ ($v=22,23 \rightarrow 0,1$) transitions in $^4\text{He}_2^+$ (uncer. 0.2-2 MHz) [2]

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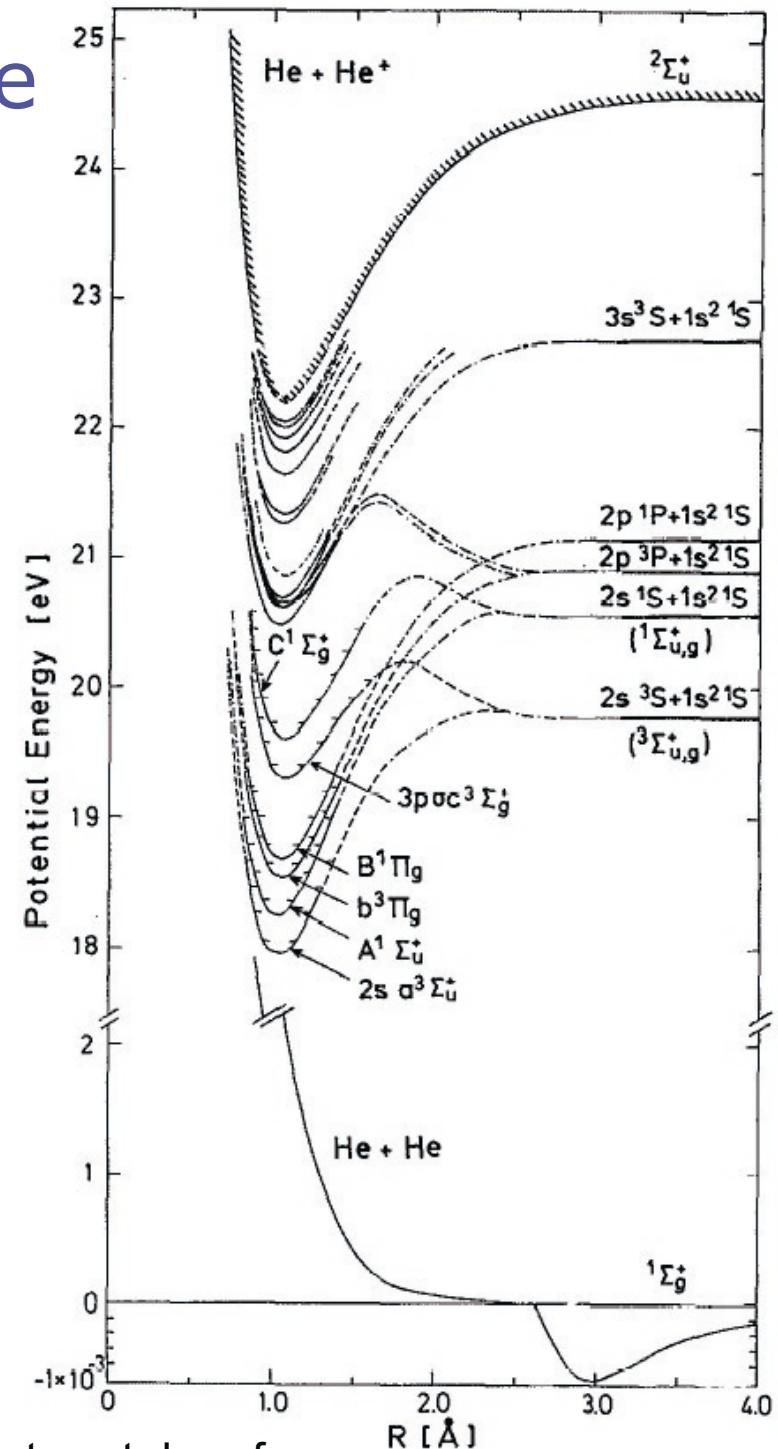
Picture taken from:

[Buchenau *et al.* JCP **95**, 8134 (1991)]

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- He_2 is a Rydberg molecule according to G. Herzberg [3]:

A molecule which has an essentially repulsive ground state and all excited states are Rydberg states



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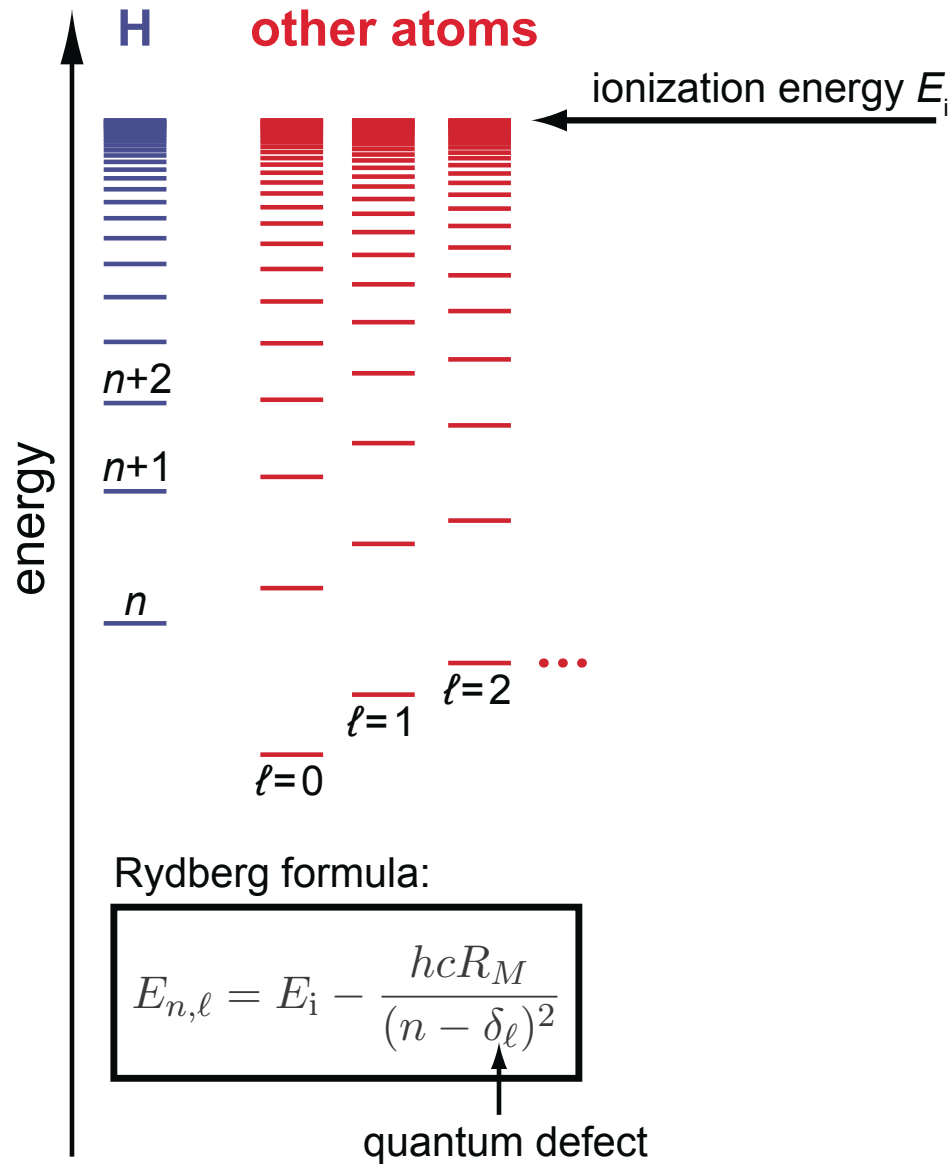
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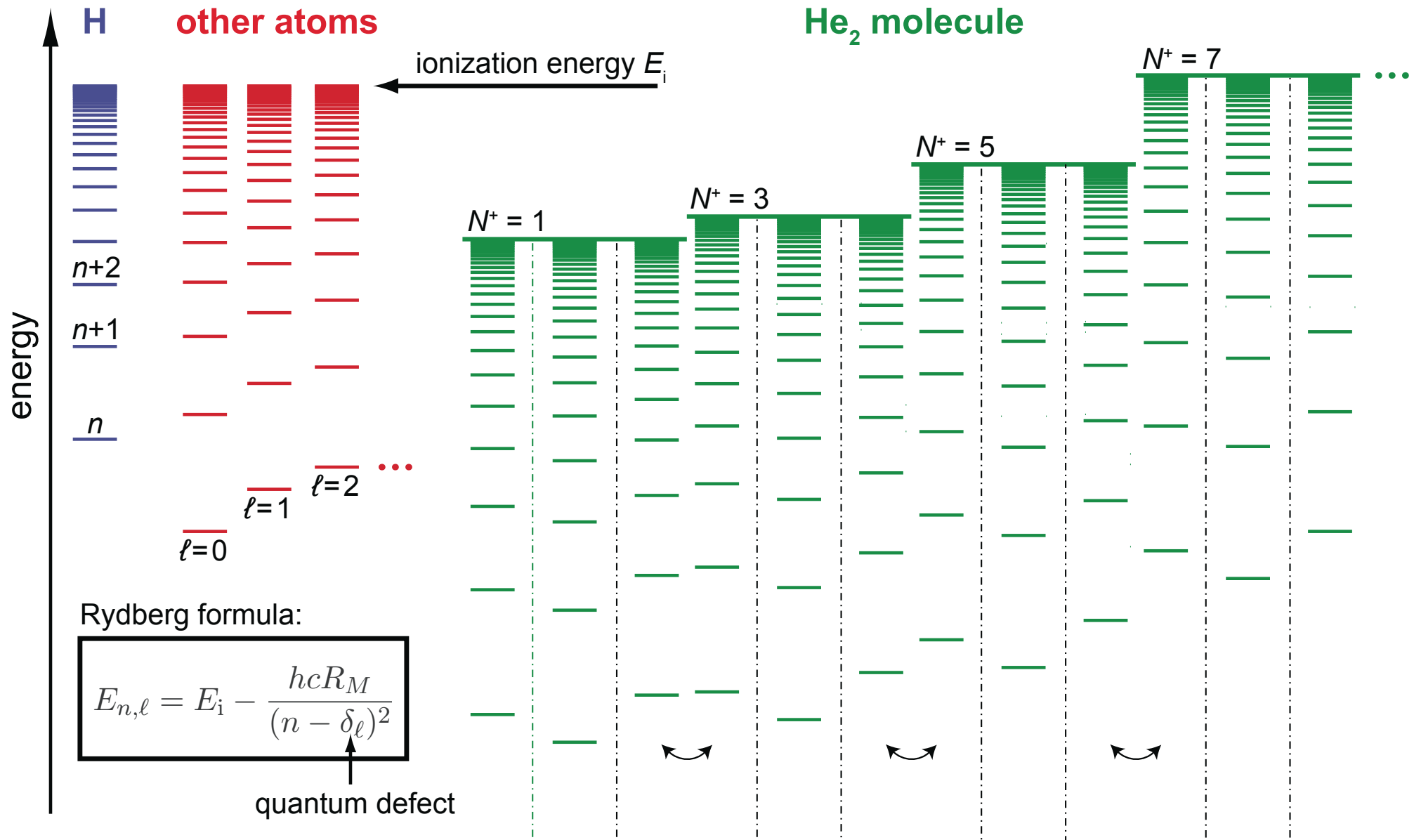
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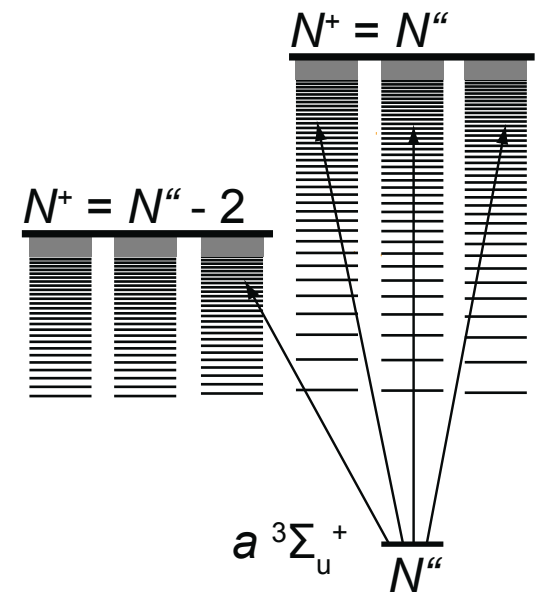
Rydberg states of atoms and molecules



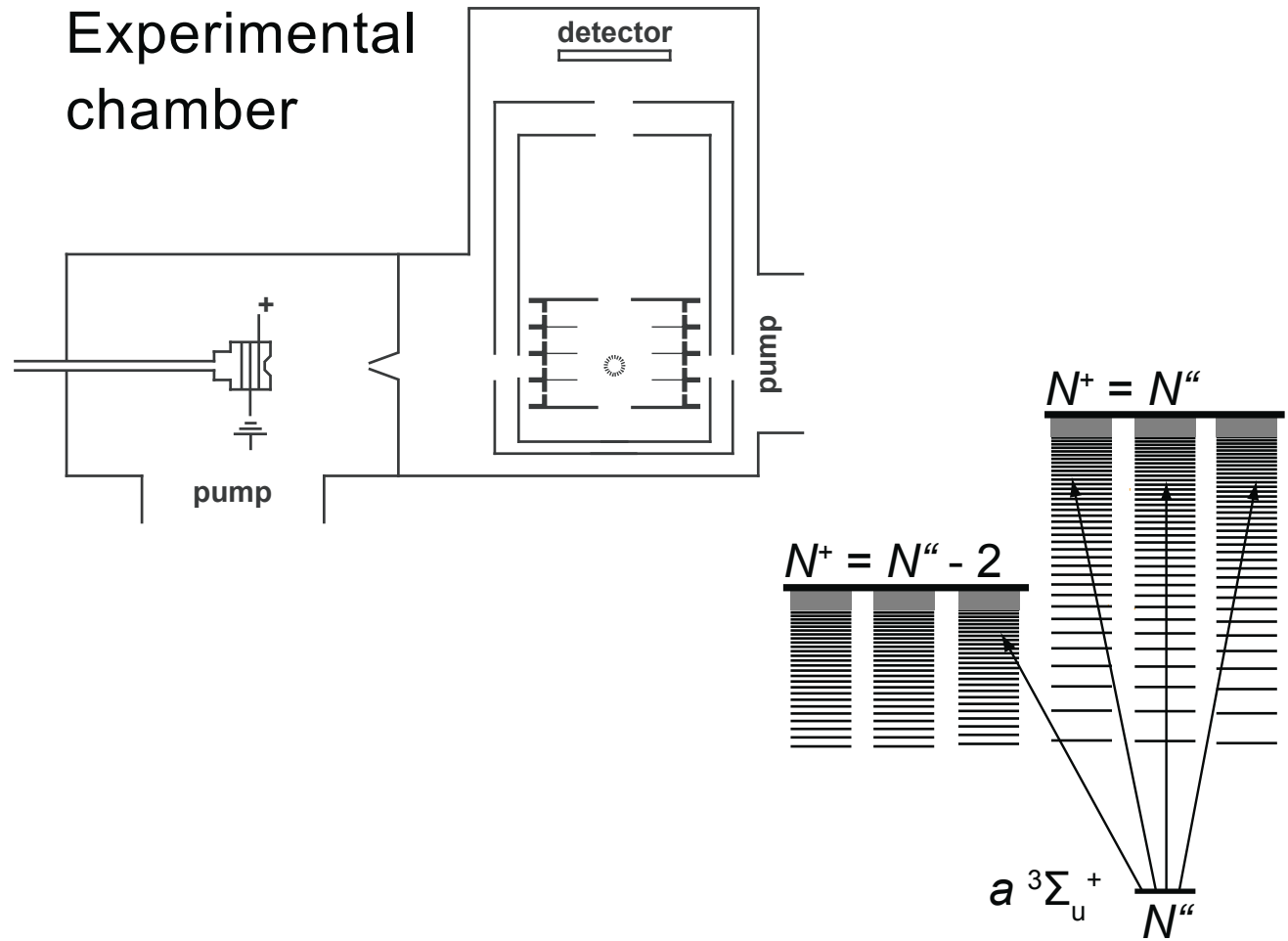
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Experimental

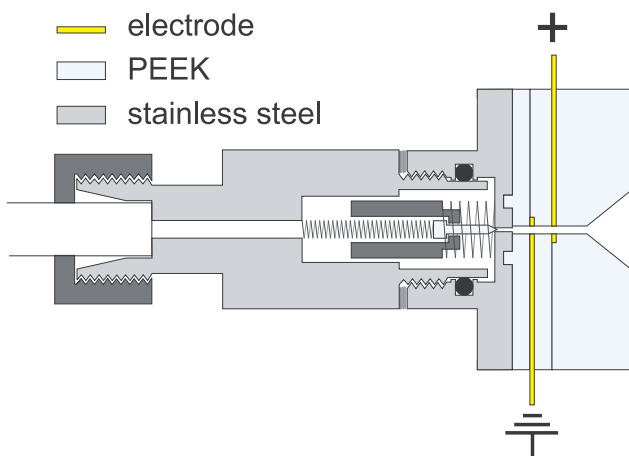


Experimental

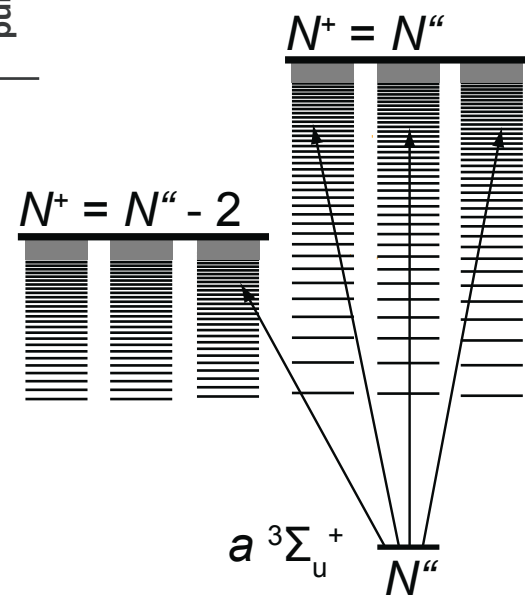
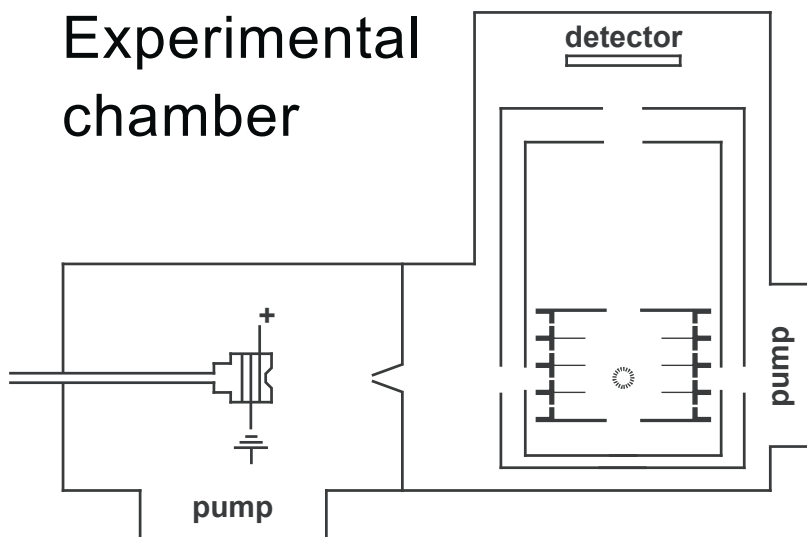


Experimental

Nozzle and discharge

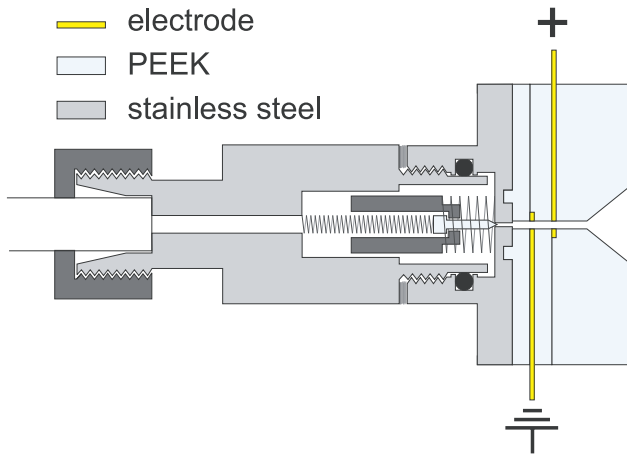


Experimental chamber

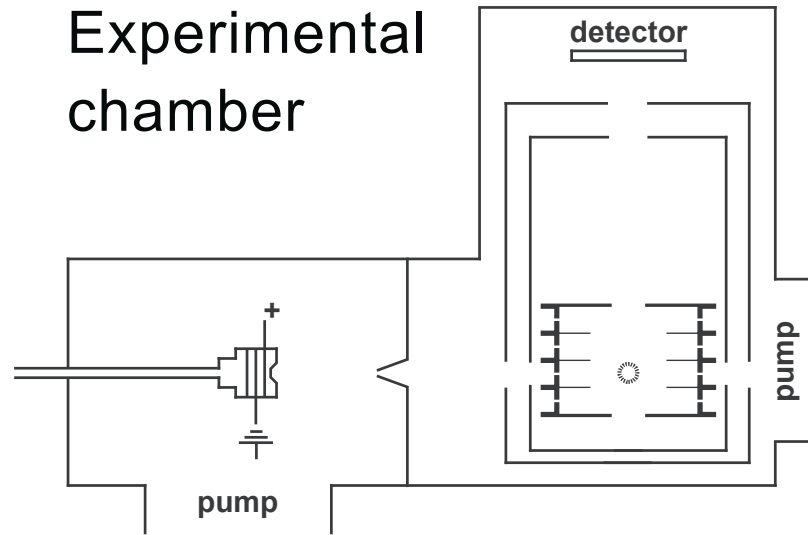


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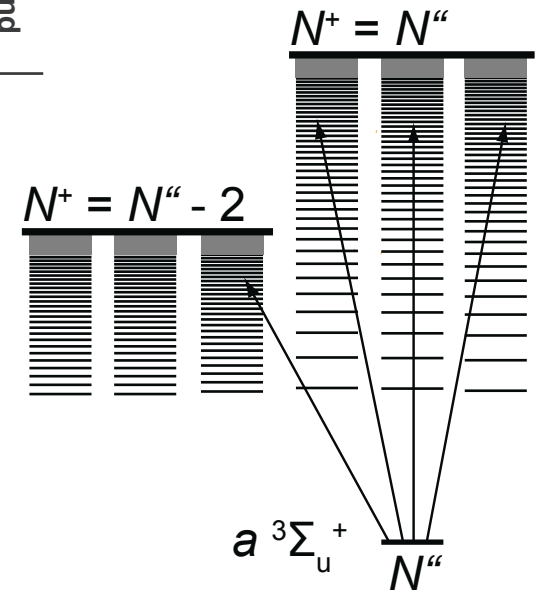
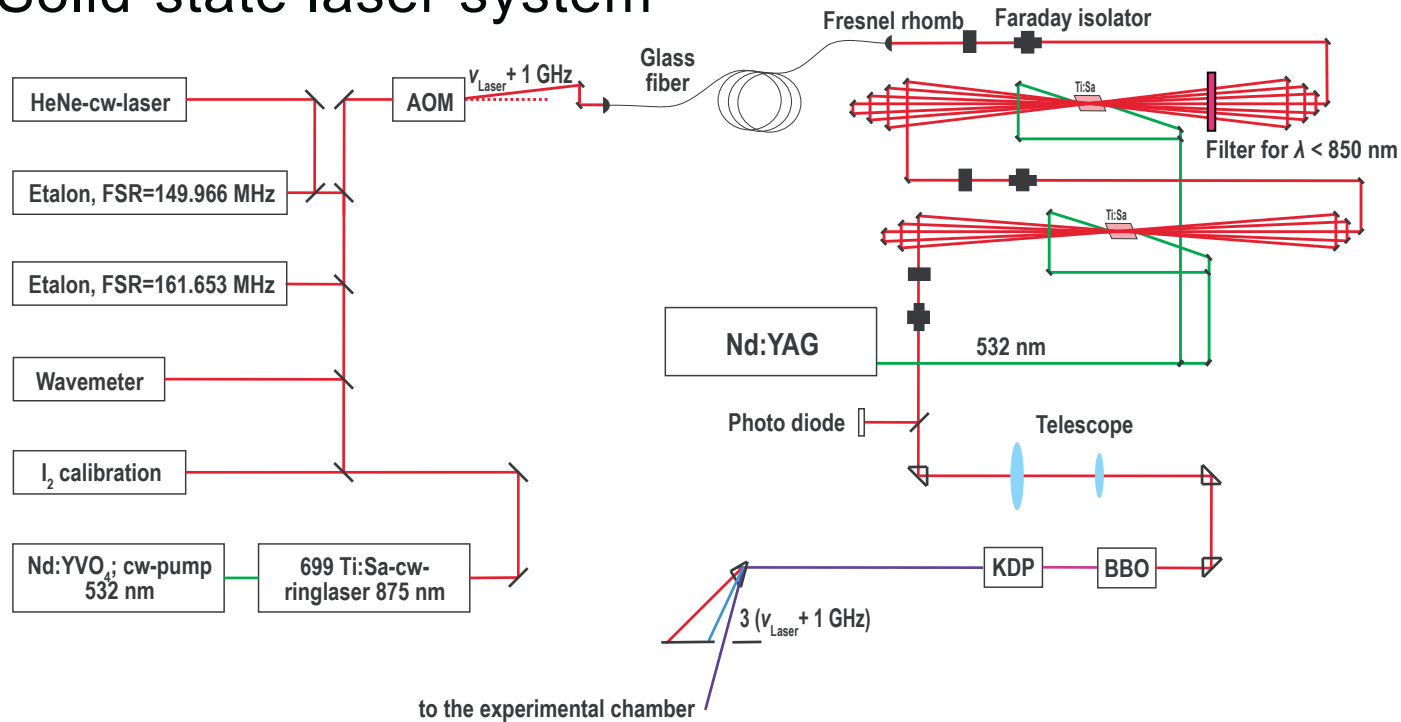
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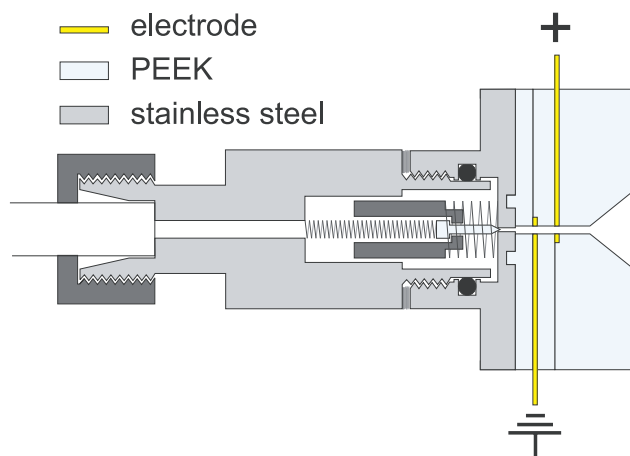


Solid-state laser system

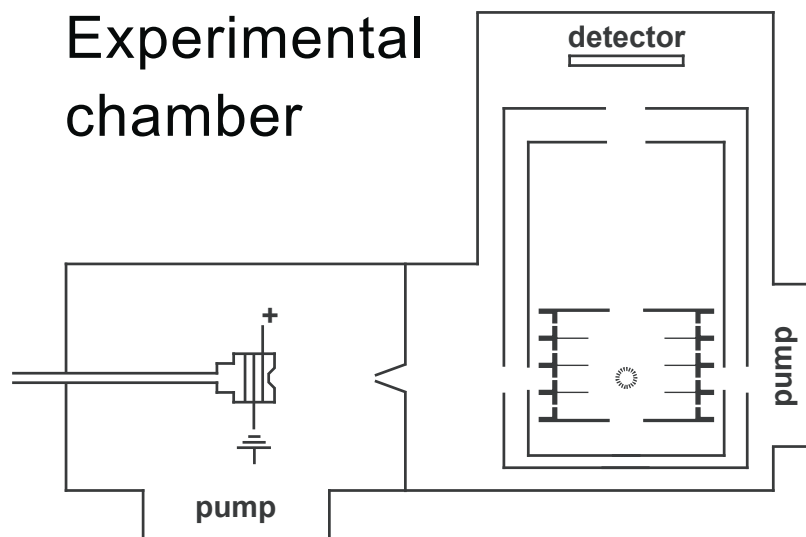


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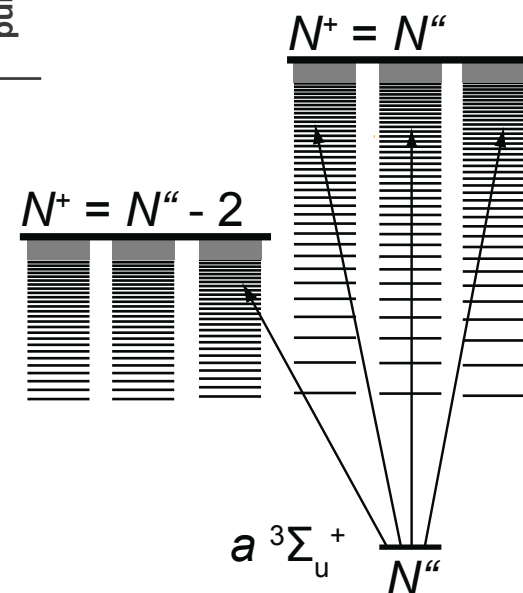
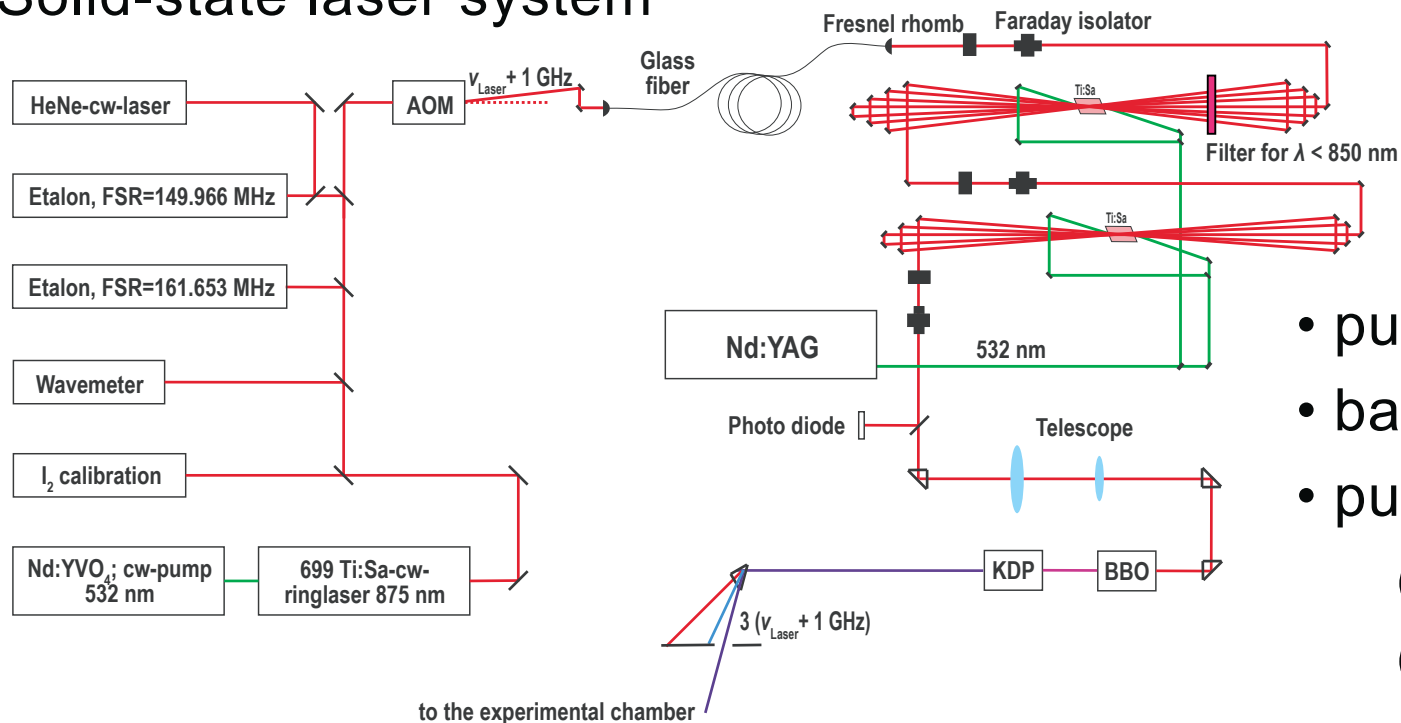
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Solid-state laser system



- pulse length ~ 40 ns
- bandwidth ~ 20 MHz
- pulse energies

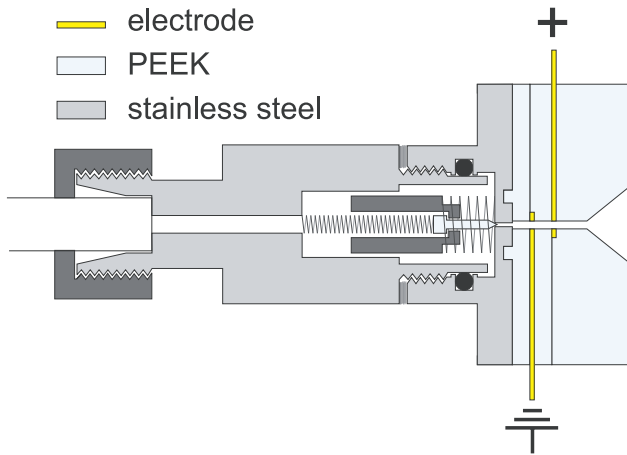
@875nm: ~ 15 mJ

@438nm: ~ 2 mJ

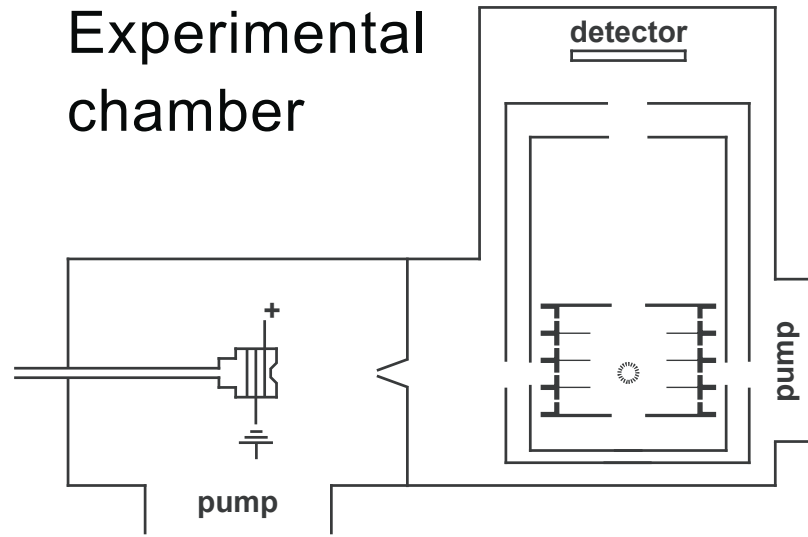
@292nm: ~ 150 μ J

Experimental

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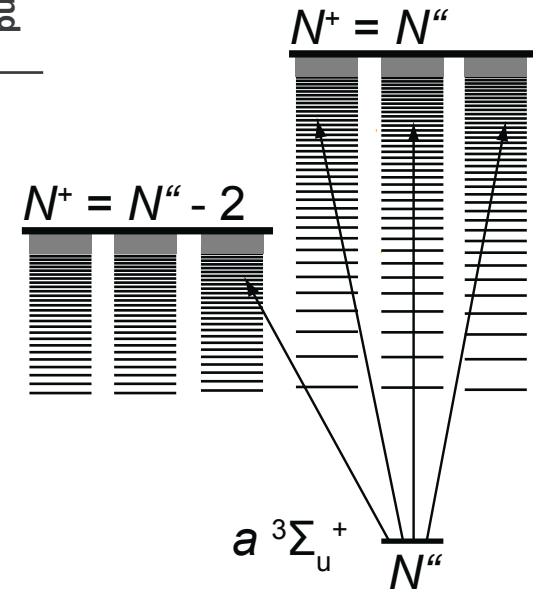
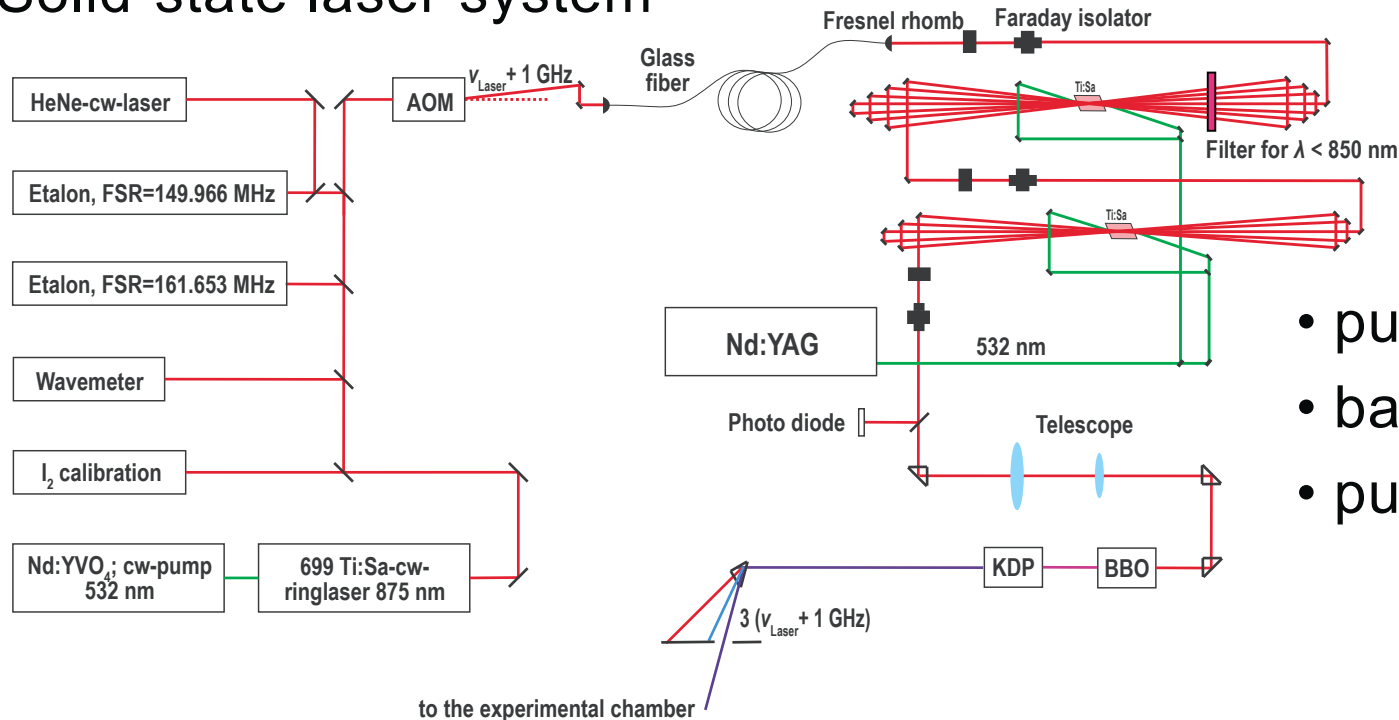


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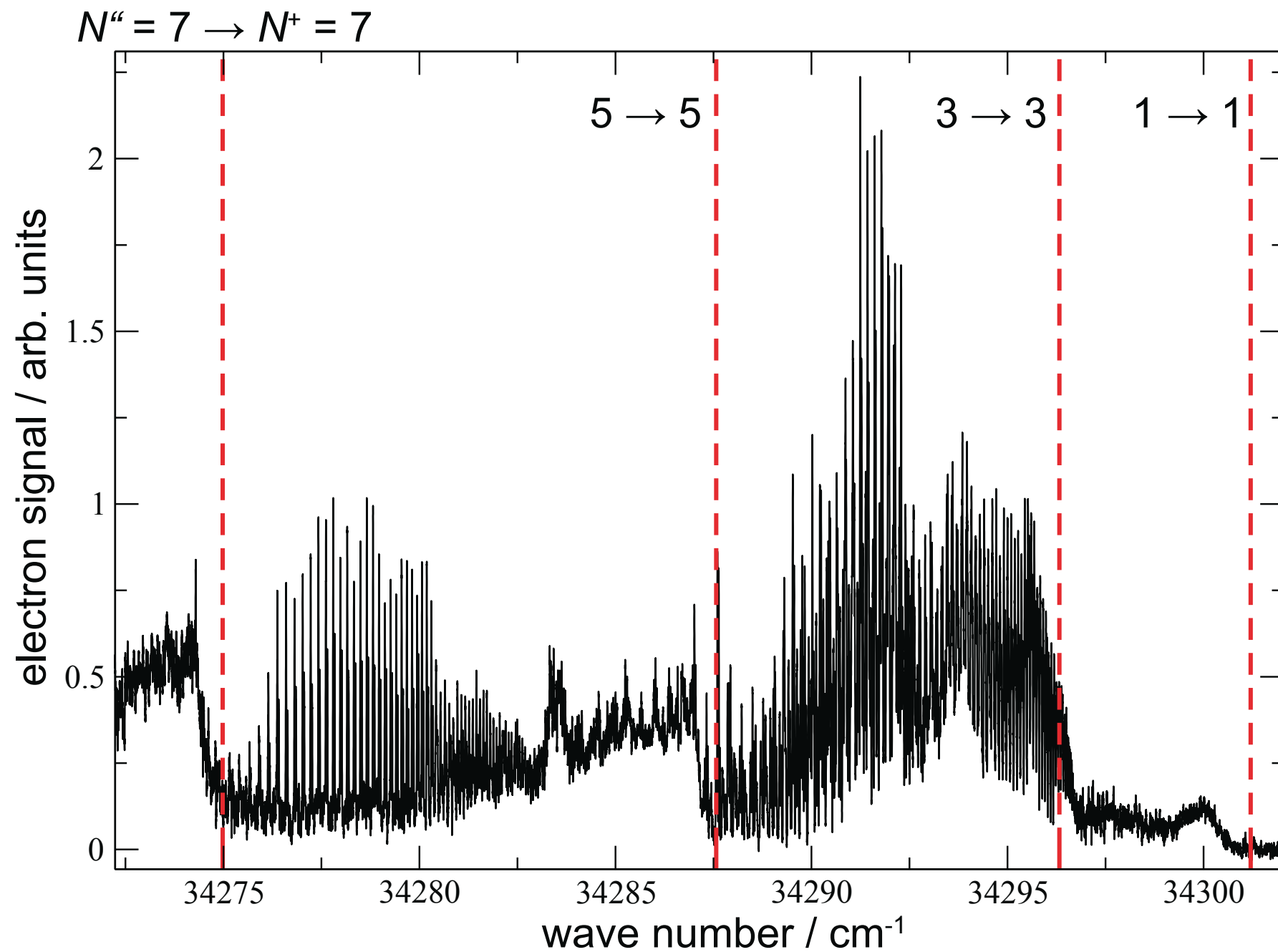
- field delay:
2 μ s
- field strength:
12.8 V/cm

Solid-state laser system

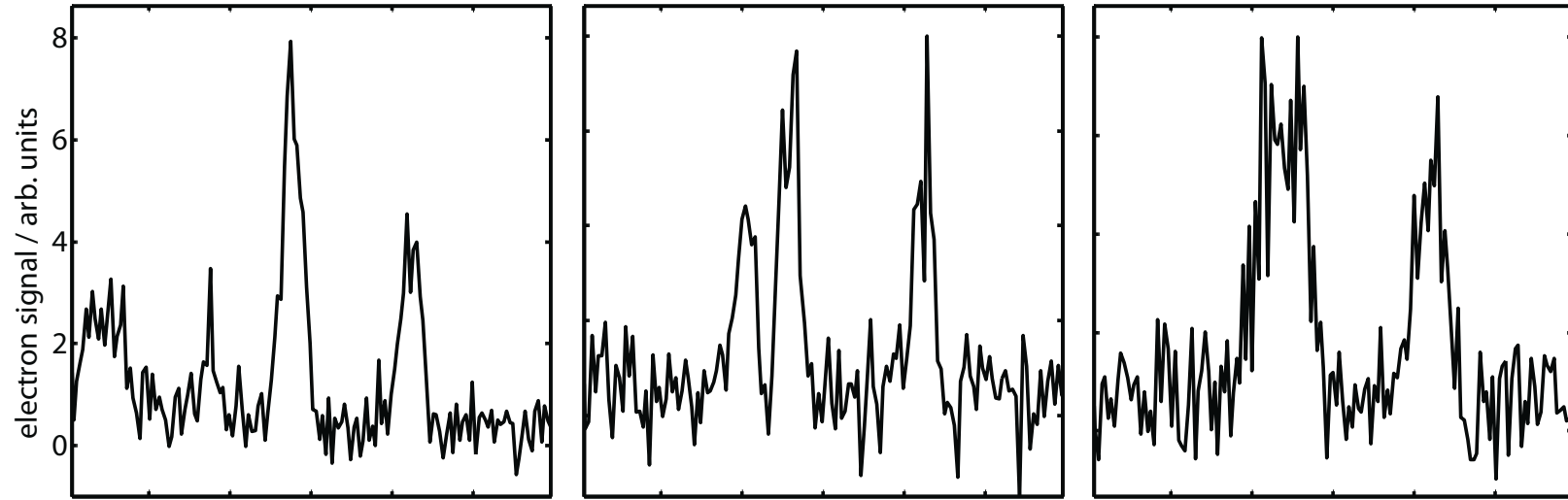


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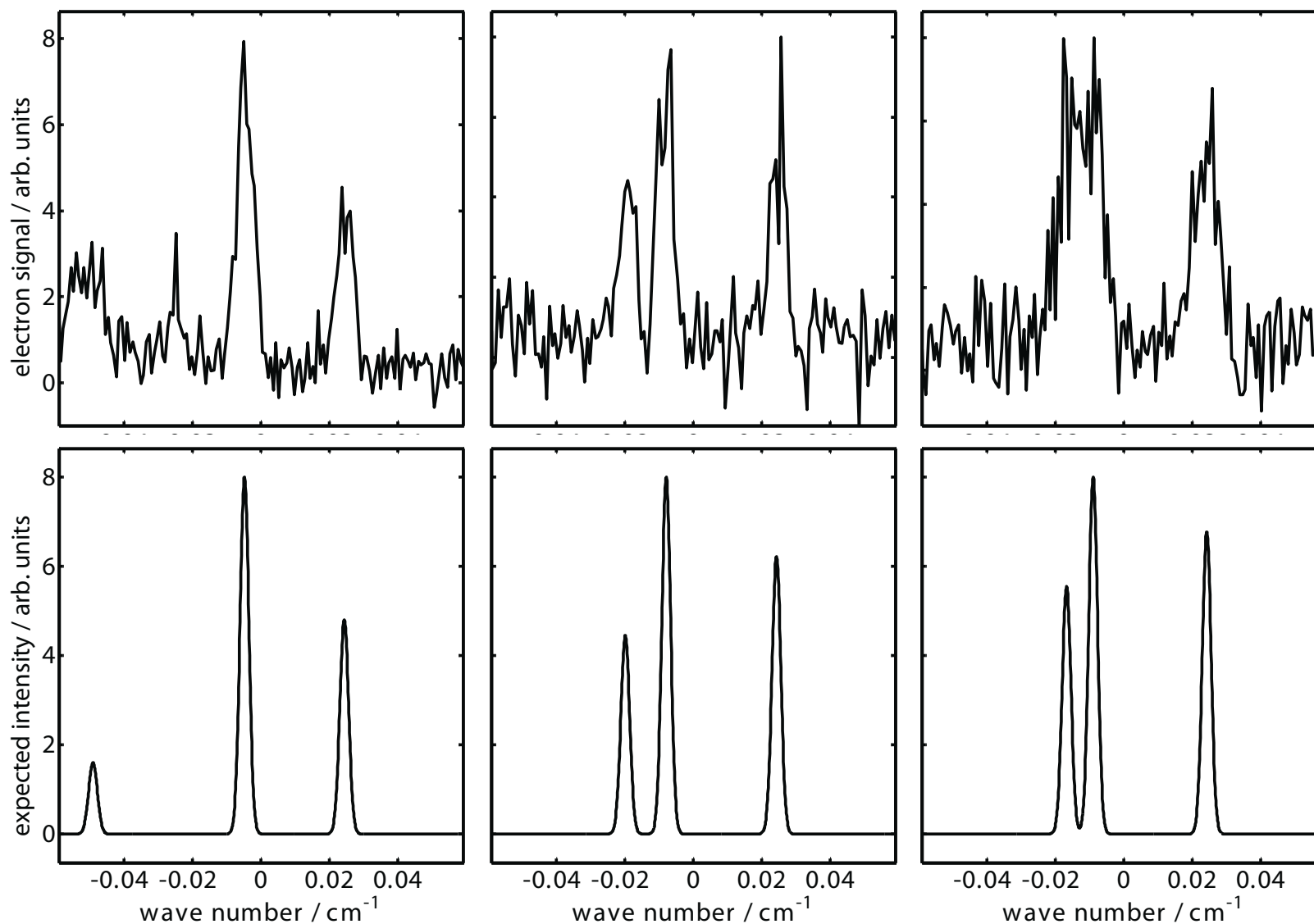
Overview Q-region



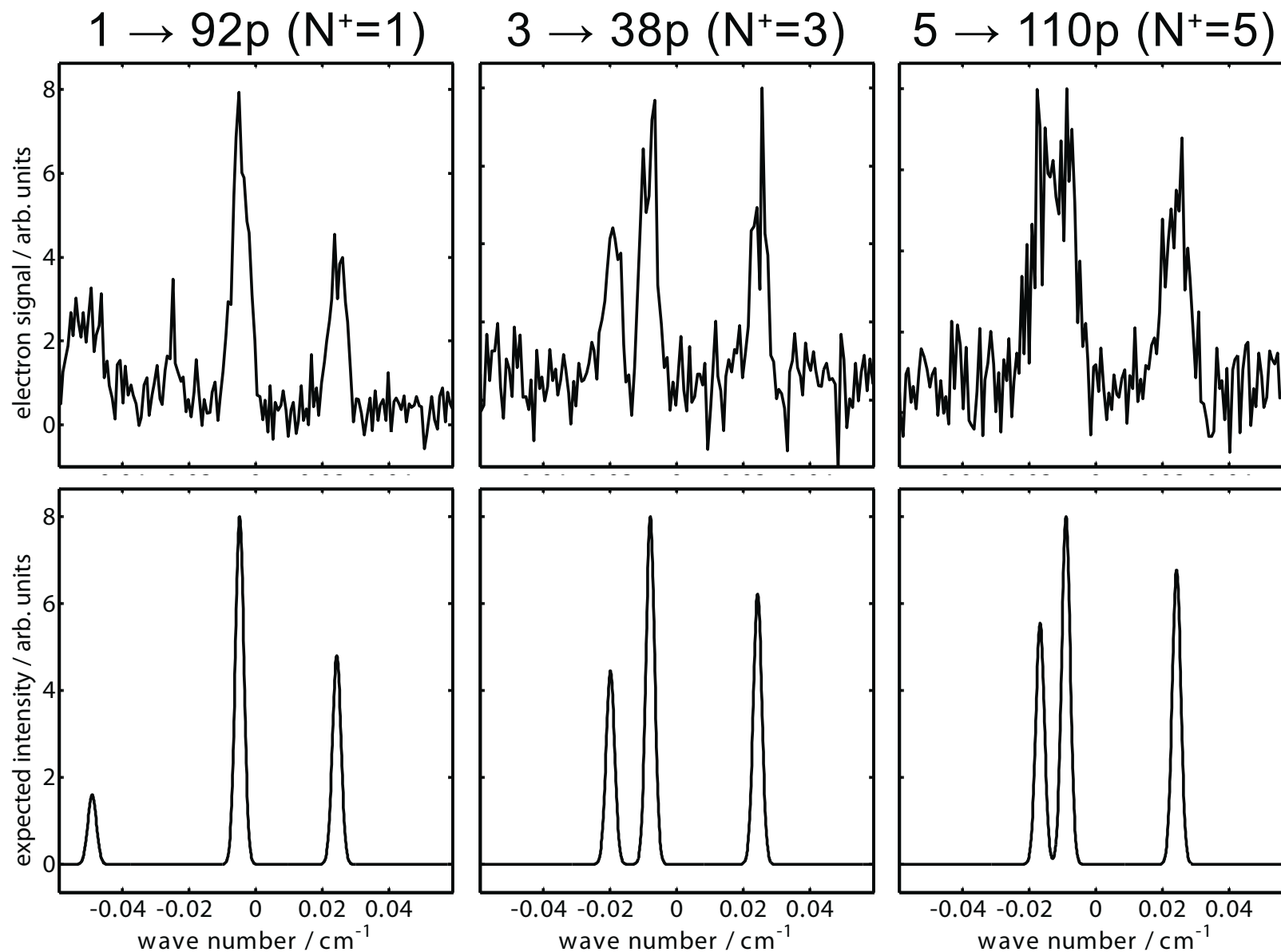
Resolved fine structure of the triplet a state



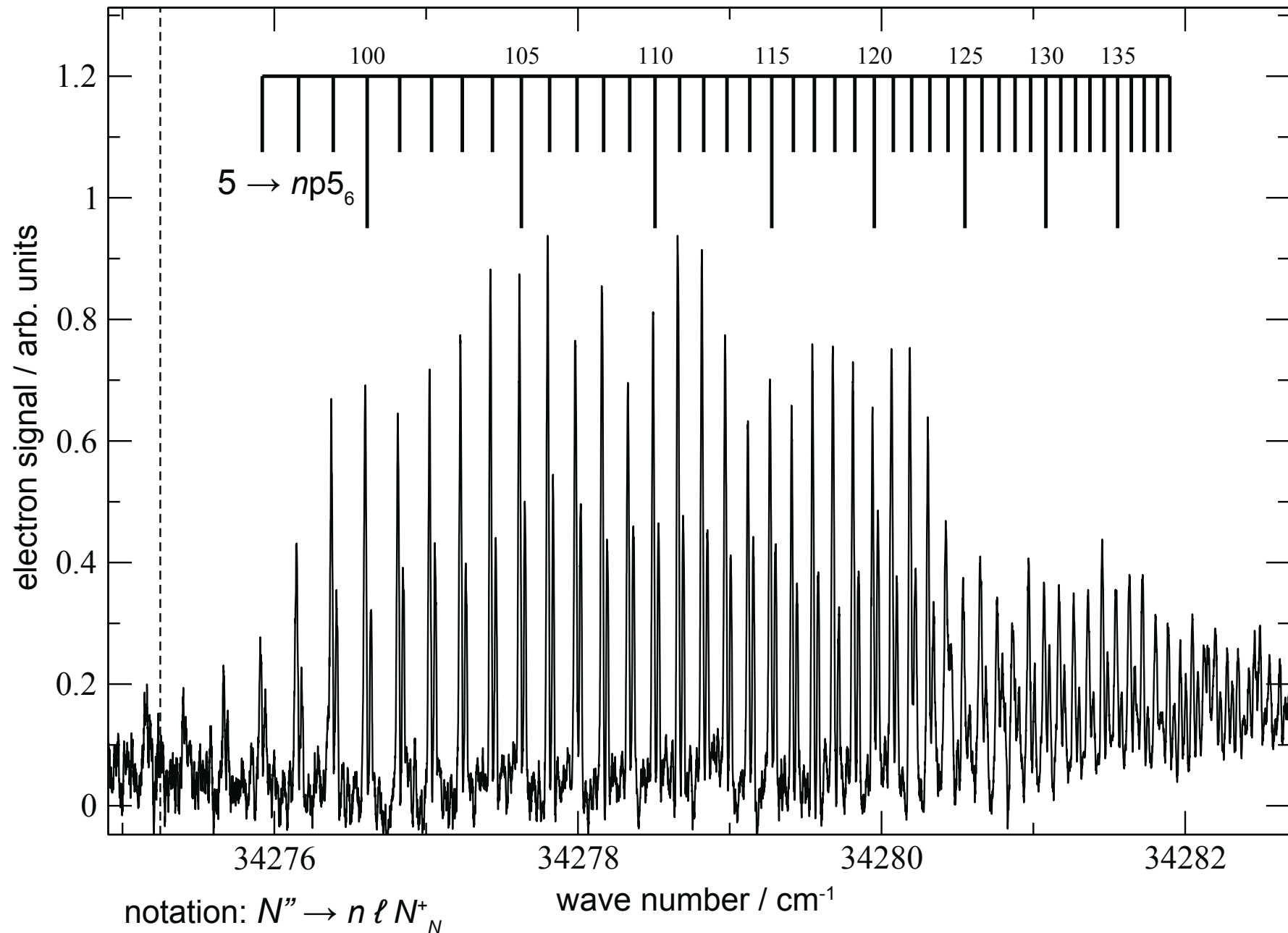
Resolved fine structure of the triplet *a* state



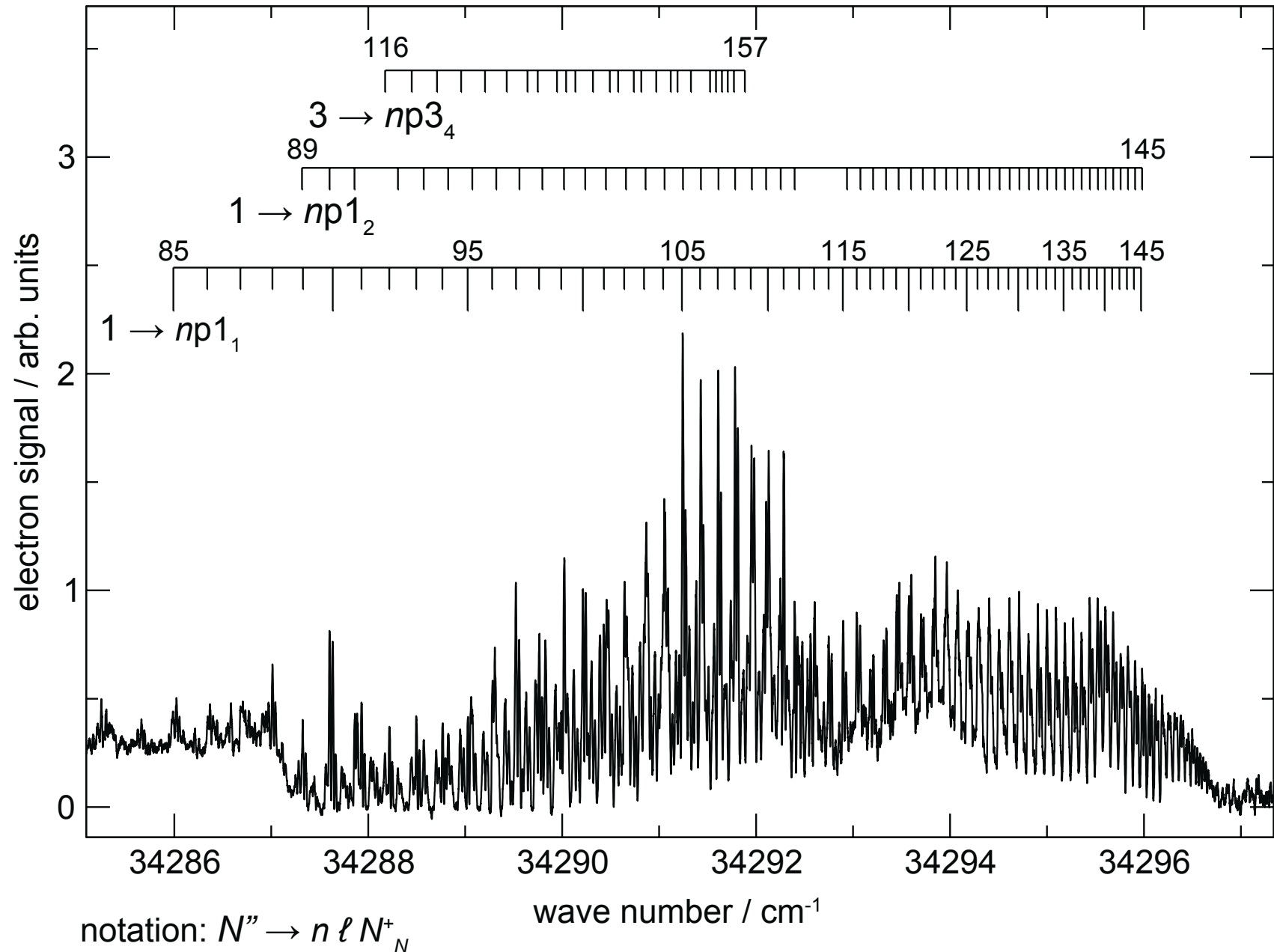
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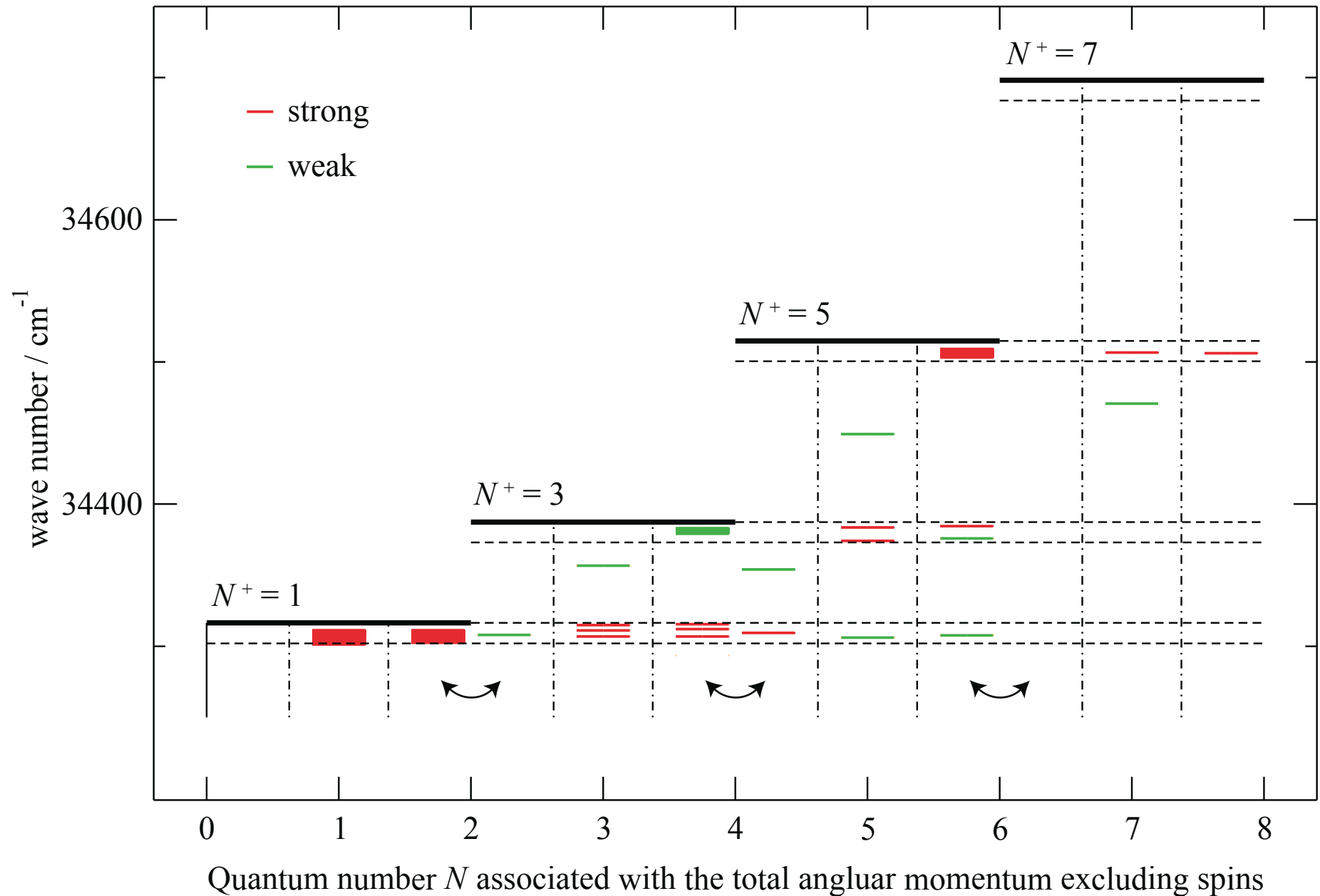
The $N^+=5$ Rydberg series



The $N^+=1,3$ Rydberg series



Overview over all observed states



Extrapolation of Rydberg series with multichannel quantum defect theory (MQDT)

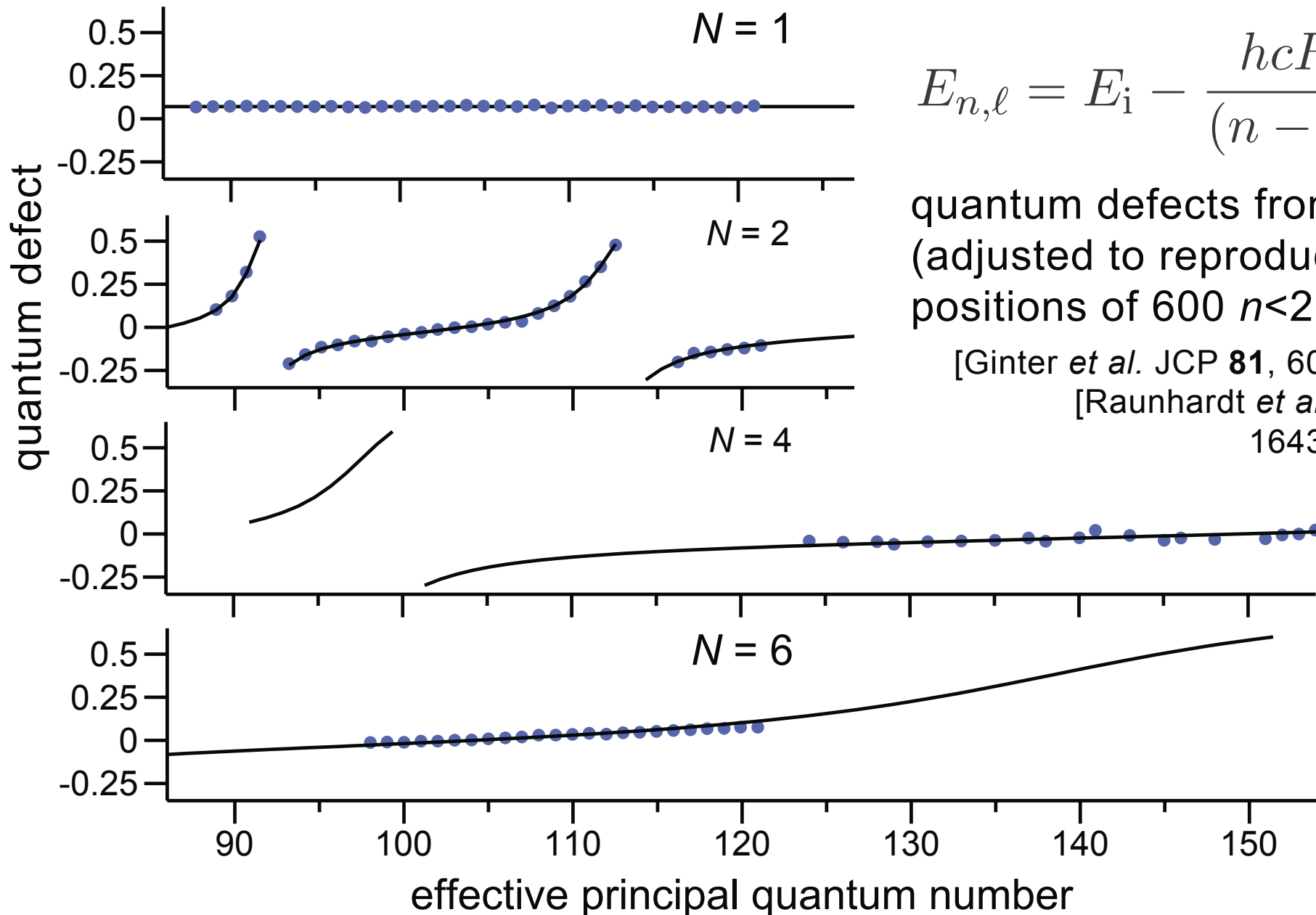
$$E_{n,\ell} = E_i - \frac{hcR_M}{(n - \delta_\ell)^2}$$

quantum defects from MQDT
(adjusted to reproduce
positions of 600 $n < 25$ states)

[Ginter *et al.* JCP **81**, 6013 (1984)]

[Raunhardt *et al.* JCP **128**,
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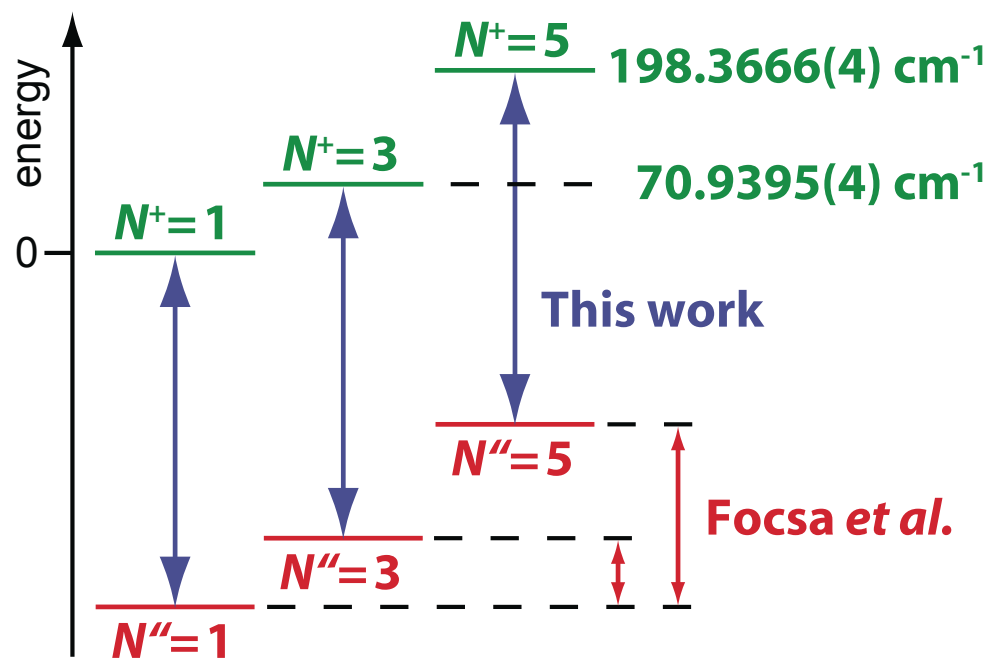
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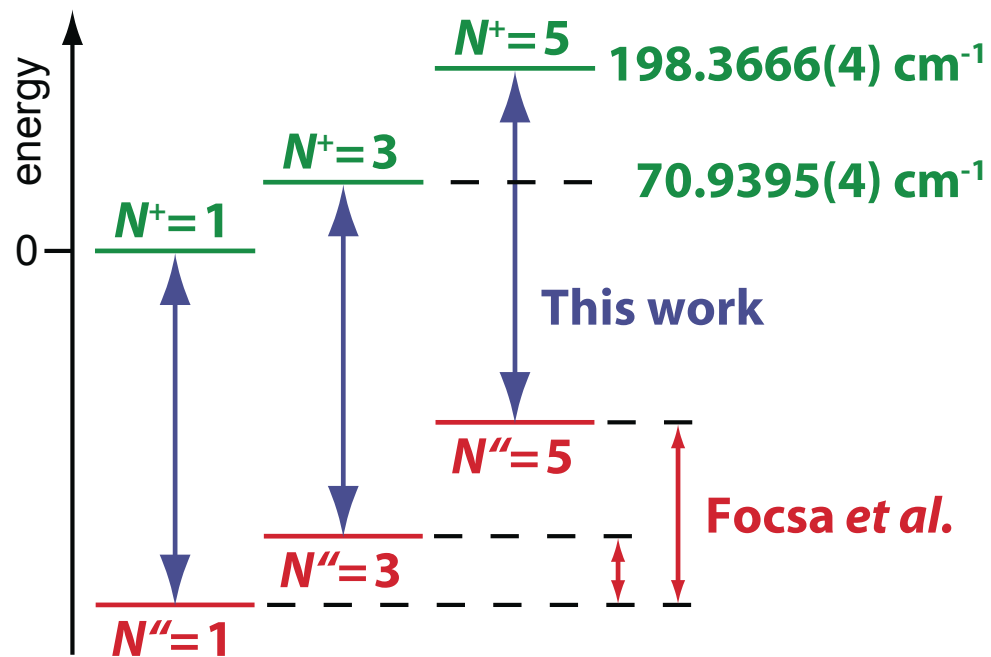
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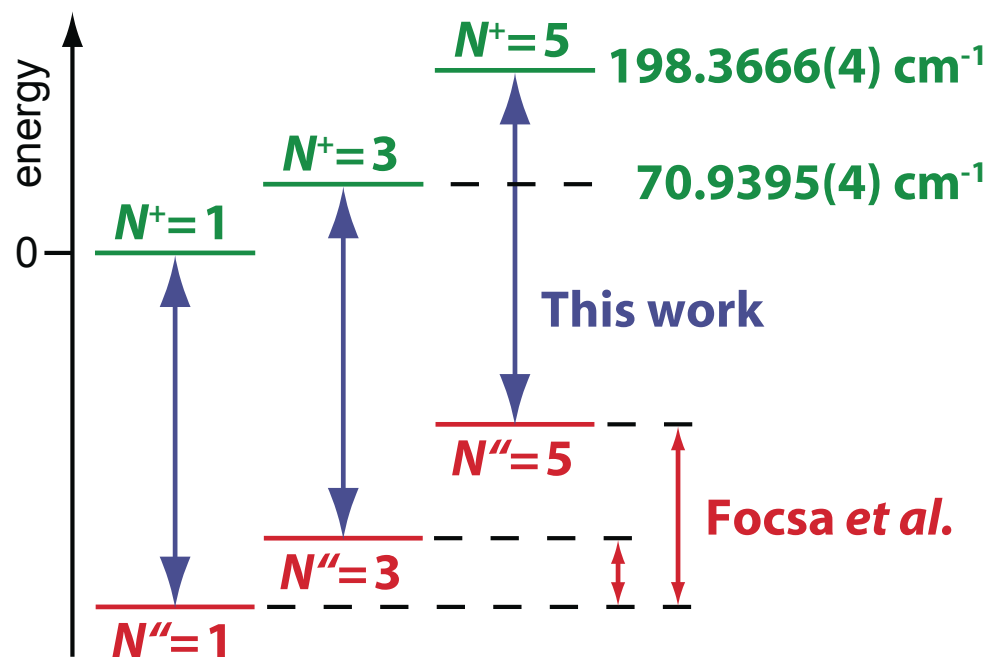


all values in cm^{-1}

N^+	Exp.	Calc. [1]	Δ
5	198.3666(4)	198.39	-0.02
3	70.9395(4)	70.95	-0.01

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Conclusions

- Transitions to triplet np Rydberg states (n up to 150) of He_2 have been resolved.
- The ionization energy of the $a\ ^3\Sigma_u^+$ state of He_2 was determined with an uncertainty of 0.006 cm⁻¹ (180 MHz).
- The energy spacing between the first three rotational states of He_2^+ could be extracted with an accuracy of 0.0004 cm⁻¹ (12 MHz).



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ETH Zurich
Switzerland
(March 2009)

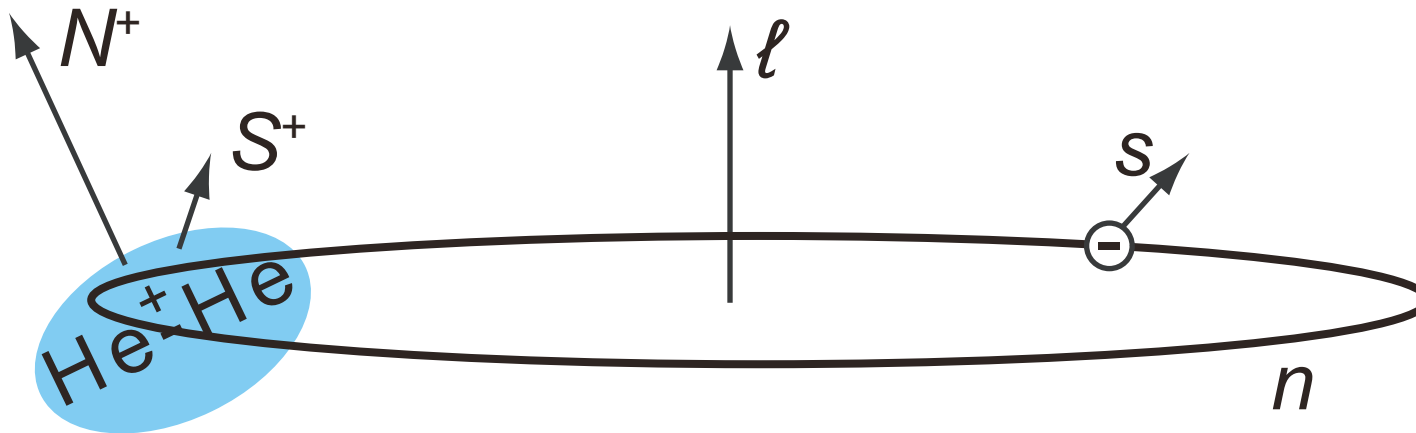




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Molecular Rydberg states and MQDT

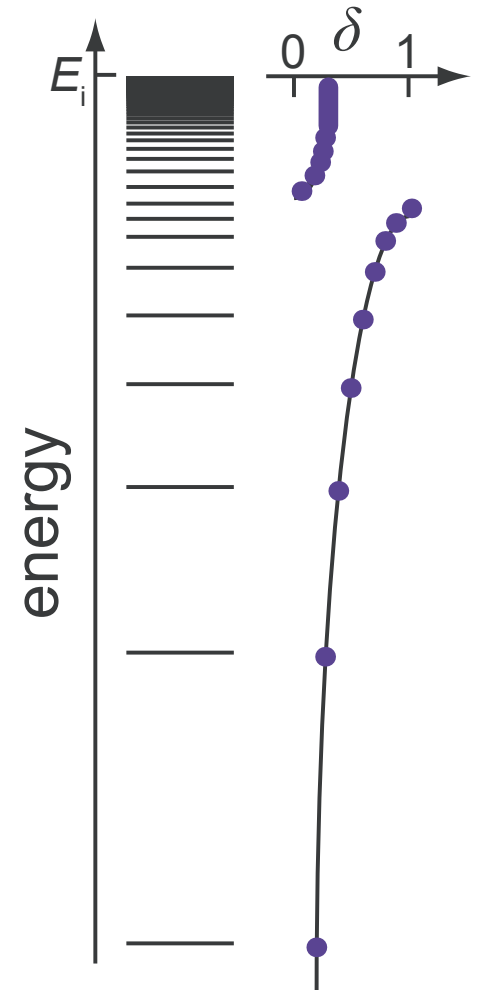


Eigen quantum defect $\mu_{\ell, \ell'}^{S, \Lambda}$

Quantization condition:

$$\det \left| U_{i\alpha} \sin[\pi(\mu_\alpha + \nu_i)] \right| = 0$$

with $\nu_i = n - \delta_i$



Triplet pp eigen quantum defects have been adjusted to reproduce positions of almost 600 $n < 25$ states:

$$E_n = E_i - \frac{hcR_N}{(n - \delta)}$$

[D.S. Ginter *et al.* JCP **81**, 6013 (1984)]
 [M. Raunhardt *et al.* JCP **128**, 164310 (2008)]

HeNe stabilized etalon

